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Standard Test Method for Alpha Particle Radioactivity of Water¹

This standard is issued under the fixed designation D 1943; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

This standard has been approved for use by agencies of the Department of Defense. Consult the DoD Index of Specifications and Standards for the specific year of issue which has been adopted by the Department of Defense.

1. Scope

1.1 This test method covers the measurement of alpha particle activity of water. It is applicable to alpha emitters having maximum energies above 3.9 MeV and at activity levels above 0.02 Bq/mL of radioactive homogeneous water. This test method is not applicable to samples containing alpha-emitting radionuclides that are volatile under conditions of the analysis.

1.2 This test method can be used for either absolute or relative determinations. In tracer work, the results may be expressed by comparison with a standard that is defined to be 100 %. For radioassay, data may be expressed in terms of alpha disintegration rates after calibration with a suitable standard. General information on radioactivity and measurement of radiation has been published² and summarized in Practice D 3648.

1.3 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

2. Referenced Documents

2.1 ASTM Standards:

D 1129 Terminology Relating to Water³

D 1193 Specification for Reagent Water³

D 2777 Practice for Determination of Precision and Bias of Applicable Methods of Committee D-19 on Water³

D 3370 Practices for Sampling Water³

¹ This test method is under the jurisdiction of ASTM Committee D-19 on Water and is the direct responsibility of Subcommittee D19.04 on Methods of Radiochemical Analysis.

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² Friedlander, G., et al., *Nuclear and Radiochemistry*, 3rd Ed., John Wiley and Sons, Inc., New York, NY, 1981.

Price, W. J., *Nuclear Radiation Detection*, 2nd Ed., McGraw-Hill Book Co., Inc., New York, NY, 1964.

Lapp, R. E., and Andrews, H. L., *Nuclear Radiation Physics*, 4th Ed., Prentice-Hall Inc., New York, NY, 1972.

Overman, R. T., and Clark, H. M., *Radioisotope Techniques*, McGraw-Hill Book Co., Inc., New York, NY, 1960.

³ *Annual Book of ASTM Standards*, Vol 11.01.

D 3648 Practice for the Measurement of Radioactivity⁴

3. Terminology

3.1 *Definitions*—For definitions of terms used in this test method, refer to Terminology D 1129. For terms not defined in this test method or in Terminology D 1129, reference may be made to other published glossaries.⁵

4. Summary of Test Method

4.1 The test sample is reduced by evaporation or a suitable chemical method to the minimum weight of material having measurable alpha activity. Alpha radioactivity is measured by an instrument composed of a detecting device, amplifier, power supply, and scaler—the most widely used being proportional and scintillation counters. In the proportional counter, which may be of the windowless or thin window type, alpha particles entering the sensitive region of the detector produce ionization of the counting gas. The negative ion of the original ion pair is accelerated towards the anode, producing additional ionization of the counting gas and developing a voltage pulse at the anode. In the scintillation detector, alpha particles interact with the material of the phosphor, transferring some of their energy to electrons. These electrons subsequently lose part of their energy by excitation rather than ionization of atoms, and the excited atoms revert to the ground state by re-emitting energy in the form of light quanta. A suitable light-sensitive device, usually a multiplier phototube, transforms the resulting flashes of light into voltage impulses. By use of suitable electronic apparatus, the pulse is amplified to a voltage sufficient for operation of the counting scaler. The number of pulses per unit time is related to the disintegration rate of the test sample. The efficiency of the system can be determined by use of a suitable alpha standard having equivalent residual plated solids.

5. Significance and Use

5.1 This test method was developed for the purpose of measuring gross alpha radioactivity in water. It is used for the analysis of both process and environmental water to determine

⁴ *Annual Book of ASTM Standards*, Vol 11.02.

⁵ American National Standard Glossary of Terms in Nuclear Science and Technology (ANSI N1.1) available from the American National Standards Institute, 1430 Broadway, New York, NY 10018.

gross alpha activity which is often a result of natural radioactivity present in minerals.

6. Measurement Variables

6.1 The relatively high absorption of alpha particles in the sample media affects the counting rate of the measurement. Effects of geometry, back-scatter, source diameter, as well as the purity, pressure variation, and type of counting gas used shall also be considered. Thus, for reliable relative measurements, the variables shall be held constant while counting all test samples and standards. For absolute measurements, appropriate efficiency factors shall be applied. If a windowless proportional counter is employed, the sample mount shall be electrically conducting.

6.1.1 In tracer studies or tests requiring only relative measurements, in which the data are expressed as being equivalent to a defined standard, the above correction factors can be simply combined into a counting efficiency factor. The use of a counting efficiency factor requires that sample mounting, material of mounting dish, and weight of residue (milligrams per square centimetre), in addition to conditions affecting the above described factors, remain constant throughout the duration of the test and that the comparative standard be prepared for counting in the same manner as the test samples. The data from comparative studies between independent laboratories when not expressed in absolute units are more meaningful when expressed as percentage relationships or as equivalent of a defined standard.

6.2 The limit of sensitivity for both scintillation and proportional counters is a function of the background counting rate which should be as low as is feasible. Massive shielding is not used for alpha counters. The maximum activity for this test method is 1600 Bq.

7. Interferences

7.1 Solids content in the sample containing the alpha emitter produces significant losses in sample counting rates of about 10 to 15 % loss at 1 mg/cm². Liquid samples shall be evaporated to dryness onto dishes that allow the sample to be counted directly by the detector. Solids on the dish shall remain constant in amount between related test samples, and should duplicate the density of the solids of the plated standard.

7.2 Most alpha counters are insensitive to beta, gamma, and X radiations.²

8. Apparatus

8.1 *Alpha Particle Counter*, consisting of either a proportional detector or a scintillation detector, and a scaler conforming to the following requirements:

8.1.1 *Proportional Detector*—This may be one of several types commercially available. The material used in the construction of the detector should contain a minimal amount of detectable radioactivity. To establish freedom from undesirable characteristics, the manufacturer shall supply voltage plateau and background counting rate data. Voltage plateau data shall show the threshold voltage, slope, and length of plateau for a particular input sensitivity.

8.1.2 *Scintillation Detector*—This may be one of several types commercially available. It shall consist of an "activated"

zinc sulfide phosphor having a minimum effective diameter of 36.5 mm and a superficial density of 10 to 15 mg/cm². The phosphor shall be mounted so that it can be attached and optically coupled to a multiplier phototube. Extraneous light shall be excluded from the phosphor either by its being covered with a thin (less than 1 mg/cm²) opaque window or by enclosing the assembly in a lightproof sample changer. The material used in the construction of the detector shall be free from detectable radioactivity. To establish freedom from undesirable characteristics, the manufacturer shall supply voltage plateau and background counting rate data. Voltage plateau data shall show the threshold voltage, slope, and length of a plateau for a specified scaler sensitivity.

8.1.3 *Scaler*—Often the scaler, mechanical register, power supply, and amplifier are contained in a single chassis, generally termed the scaler. The power supply and amplifier sections shall be matched with the type of detector to produce satisfactory operating characteristics and to provide sufficient range in adjustments to maintain controlled conditions. The manufacturer shall provide resolving time information for the counting system. The scaler shall have capacity for storing and visually displaying at least 10⁶ counts with a resolving time no greater than 5 μ s. The instrument shall have an adjustable input sensitivity that can be matched to the detector and a variable high voltage power supply with indicating meter.

8.2 *Sample Mounting Dish*—Dishes having a flat bottom of a diameter slightly less than the inside diameter of the detector. Flat dishes are preferred, but dishes may be used that have 3.2-mm high side walls with the angle between dish bottom and side equal to or greater than 120°. Dishes shall be of a material that will not corrode under the plating conditions and shall be of uniform surface density; platinum and stainless steel have been used for this purpose.

9. Reagents

9.1 *Purity of Reagents*—Reagent grade chemicals shall be used in all tests. Unless otherwise indicated, it is intended that all reagents shall conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society, where such specifications are available.⁶ Other grades may be used, provided it is first ascertained that the reagent is of sufficiently high purity and free from radioactivity to preclude detrimental effects. Some chemicals, even of high purity, contain naturally occurring radioactive elements, for example, uranium, actinium, and thorium. Consequently, when carrier chemicals are used in the analysis of low-radioactivity samples, the radioactivity of the carriers shall be determined under identical analytical conditions of the sample including residual dish solids. The radioactivity of the reagents shall be considered as background and subtracted from the test sample counting rate.

9.2 *Purity of Water*—Unless otherwise indicated, references to water shall be understood to mean reagent water conforming

⁶ *Reagent Chemicals, American Chemical Society Specifications*, American Chemical Society, Washington, DC. For suggestions on the testing of reagents not listed by the American Chemical Society, see *Analar Standards for Laboratory Chemicals*, BDH Ltd., Poole, Dorset, U.K., and the *United States Pharmacopeia and National Formulary*, U.S. Pharmaceutical Convention, Inc. (USPC), Rockville, MD.

to Specification D 1193, Type HI.

9.3 *Nitric Acid* (sp gr 1.42)—Concentrated nitric acid (HNO_3).

9.4 *Nitric Acid* (1 + 30)—Mix 1 volume of concentrated HNO_3 (sp gr 1.42) with 30 volumes of water.

9.5 *Alpha-Emitting Radioactive Standard Solution* (~200 Bq/mL), traceable to the National Institute of Standards and Technology (NIST).

10. Sampling

10.1 Collect the sample in accordance with Practices D 3370.

10.2 Preserve the sample in a radioactive homogeneous state. A sample shall be made radioactive homogeneous by addition of a reagent in which the radionuclides or compounds of the radionuclides present would be soluble in large concentrations. Addition of acids, complexing agents, or stable chemically similar carriers may be used to obtain homogeneity. Consideration of the chemical nature of the radionuclides and compounds present and the subsequent chemistry of the method shall indicate the action to be taken.

11. Establishing Gas Proportional Counter Operating Plateau

11.1 Put the instrument into operation according to the manufacturer's instructions. Place a quality control (QC) source in the detector, set the sensitivity control near its maximum and turn the "count" switch to "count" position. Slowly increase the high voltage until the first counts are observed and record the "threshold" voltage. Advance the voltage in increments of convenient magnitude (approximately 25 V) and determine the counting rate at four or more settings of the sensitivity control at each voltage setting. Measure the background counting rate at each of the settings using an empty sample mounting dish in place of the standard.

11.1.1 The QC source may be any available alpha-emitting radionuclide having a half life sufficiently long to eliminate decay corrections. Knowledge of its true disintegration rate is not essential. The radionuclide shall be permanently fixed to the dish and uniformly distributed over an area preferably smaller than the dish bottom; electro-deposition and flaming of a salt-free solution are the two methods most generally used. Quality control sources are commercially available.

11.2 Plot the gross counting rate of the standard against the voltage. The counting rate should rise initially as the voltage is increased, then, for at least some of the settings of the sensitivity control, reach an approximate constant value, and finally rise again. The "plateau" of the curve should be at least 100 V in length and have a slope less than 2 %/100 V; however, shorter plateaus or one with greater slope shall be acceptable if a well regulated high voltage power supply is available.

11.3 Plot the ratio of the square of the net counting rate of the standard to the background counting rate against the voltage for each of the settings of the sensitivity control.

11.4 Determine the optimum conditions for operation of the instrument by selecting values for the high-voltage and sensitivity adjustments that correspond to some point lying on the plateau of the counting-rate-versus-voltage plot and near the maximum value of the ratio of the sample-squared-to-

background counting rates.

12. Control of Instrument Operation

12.1 Tolerance or statistical control charts are used to assure that the instrument is operating to within pre-specified limits of the initial calibration. Repetitive measurements of a quality control source are taken to develop the tolerance or statistical control chart. The QC source is then used on a daily or prior-to-use basis to ensure proper operation. Refer to Practice D 3648 for the preparation of a tolerance or statistical control chart.

13. Calibration and Standardization for General Measurements

13.1 Place a known amount of a NIST-traceable alpha standard (approximately 200 Bq) into a volume of water sufficient to dissolve salts (or into a volume of water containing dissolved salts) equivalent to those of the test samples and prepare for counting as directed in Section 15. Throughout the experiment, the evaporation, mounting, counting, and density of plate solids of this reference standard shall be identical with those of the test samples. Count for a length of time required to produce the desired statistical reliability (typically 1 %). The efficiency factor for each dissolved salt weight, f_o , is then expressed as a fraction of the disintegration rate (Bq) of the reference standard and is calculated according to the following equation:

$$f_o = \text{cps} / \text{Bq} \quad (1)$$

where:

cps = the measured counts per second.

The alpha emitting standard should have approximately the same alpha particle energy as the nuclides of interest so that mass attenuation effects can be estimated appropriately.

13.1.1 Purified natural uranium, of which the specific activity is 0.25 Bq per microgram, has been found satisfactory for this purpose. Other alpha-emitter preparations of known disintegration rate, for example, ^{241}Am or ^{237}Np , may also be used. When available, all calibration solutions shall be NIST traceable.

14. Calibration and Standardization for Tracer Experiments

14.1 Add a known quantity of activity from a reference solution of the tracer (approximately 180 Bq) to a radioactivity-free standard test sample and process as directed in Section 15.

15. Procedure

15.1 Place an appropriate volume of the test solution in a glass beaker, add 3 mL of concentrated HNO_3 (sp gr 1.42) for each 100 mL of solution, and evaporate to 1 to 2 mL. Quantitatively transfer to the mounting dish and evaporate to dryness. Adjust the heat carefully to prevent spattering or boiling. A ring heater having a continuously variable voltage control or adjustable infrared heat lamps are the preferable heat sources for the final evaporation and drying. Uniform spreading of the residual salts is necessary for reliable comparative data. After drying, heat the dish to dull redness for a few

seconds using a burner. Cool hygroscopic solids in a dry atmosphere and store in a desiccator until the start of counting. Place the sample in the counter and count for a time interval sufficient to attain the desired statistical reliability. Record the reading of the register. Transfer of large volume samples to smaller beakers as evaporation nears completion makes for easier transfer of the test specimen to the mounting dish. Make all transfers with $\text{HNO}_3(1+30)$. Choose the sample size with consideration for the absorption of alpha particles in the residual solids. The size should be such that the density of the deposit on the plate shall not exceed 5 mg/cm^2 .

15.2 Precipitation methods may be used expediently to concentrate the radioactive material into small amounts of precipitate. The precipitate is separated and washed free of precipitant by centrifugation or filtration. Choose the method of separation that will produce a uniform deposit of precipitate after quantitatively transferring to the mounting dish for counting. Calibrate the instrument under counting conditions identical to those used for the samples. More detailed information is published² on the techniques and equipment for separation and mounting of the precipitate.

16. Calculation

16.1 Results may be expressed in observed counts per second per millilitre or Bq/mL. This test method is useful for comparing activities of a group of samples, as in tracer experiments. Results may also be reported in terms of equivalent americium-241 activity or other standard radionuclide activity using the empirical efficiency determined by use of a reference standard. If it is known that only one nuclide is present, its disintegration rate may be determined by use of the efficiency factor determined from a reference standard of that nuclide obtained from the National Institute of Standards and Technology (NIST) or from a NIST-traceable standard. Calculate the results as follows:

$$\text{alpha concentration (Bq/mL)} = C_{\text{net}} / (f_o \times V) \quad (2)$$

where:

C_{net} = net count rate (s^{-1}),
 V = test specimen, mL, and
 f_o = detector efficiency factor.

The total propagated uncertainty of the alpha concentration is calculated as:

$$\sigma_{\text{Bq/mL}} = \text{Bq/mL} \times [(\sigma_{C_{\text{net}}}/C_{\text{net}})^2 + (\sigma_{f_o}/f_o)^2 + (\sigma_V/V)^2]^{1/2} \quad (3)$$

where:

$\sigma_{C_{\text{net}}}/C_{\text{net}}$ = relative counting uncertainty,
 σ_{f_o}/f_o = relative detector efficiency uncertainty, and
 σ_V/V = relative uncertainty in the sample volume measurement.

The net count rate and counting uncertainty, $\sigma_{C_{\text{net}}}$, are defined as:

$$C_{\text{net}} = CR_S - CR_B = C_S / t_S - C_B / t_B \quad (4)$$

$$\sigma_{C_{\text{net}}} = (C_S / t_S^2 + C_B / t_B^2)^{1/2} \quad (5)$$

where:

CR_S = sample count rate (s^{-1}),
 CR_B = background count rate (s^{-1}),

C_S = sample counts,
 C_B = background counts,
 t_S = counting time of sample(s), and
 t_B = counting time of background(s).

The *a priori* minimum detectable concentration (MDC) is calculated using the equation:

$$\text{MDC} = 2.71 / (t_S \times k) + 4.65 \times \sigma_B / k \quad (6)$$

where:

$\sigma_B = (CR_B / t_S)^{1/2}$, and
 $k = f_o \times V$.

A more detailed discussion on the minimum detectable concentration concept can be found in Practice D 3648.

17. Precision and Bias

17.1 The overall precision and bias of this test method within its designated range varies with the quantity being tested according to Table 1.

17.2 This collaborative test for the determination of gross alpha activity in water was conducted by six laboratories at three concentration levels ranging from 1.03 to 4.17 Bq/mL and containing 8 mg, 19.5 mg, and 40 mg of solids, respectively. Each laboratory processed three replicates per level.

17.3 The precision and bias statements for this test method were obtained using Practice D 2777 – 86.

18. Quality Control

18.1 Before this test method is utilized for the analysis of samples, a counter quality control or tolerance chart shall be established to ensure that the counting system is operating within prescribed limits. The quality control or tolerance chart shall be established at the time the counting system is calibrated.

18.2 Prepare a quality control or tolerance chart as recommended in Practice D 3648. The counting system shall be checked by analyzing a QC source daily or prior to use. The result of the QC analysis shall be tabulated or plotted on the control or tolerance chart and evaluated according to Practice D 3648.

18.3 Evaluate the counting system background periodically. The background data shall be maintained in a logbook or plotted on a trend chart.

18.4 Precision and bias can be assessed in the following manner: the precision of an individual measurement can be approximated by the total propagated uncertainty and bias can be assessed by the analysis of NIST traceable spiked samples with known quantities of radioactivity.

19. Keywords

19.1 gross alpha radioactivity; gross radioactivity measurement; proportional counter; water

TABLE 1 Determination of Precision and Bias

Amount Added, Bq/mL	Average Calculated Amount, Bq/mL	± Bias	± % Bias	Statistically Significant (5 % C.I.)	Precision	
					S_t	S_o
1.03 ± 0.01	0.98 ± 0.20	-0.05	-4.9	No	0.195	0.058
2.08 ± 0.02	2.08 ± 0.46	+ 0.00	+ 0.0	No	0.458	0.137
4.17 ± 0.05	3.97 ± 0.83	-0.20	-4.8	No	0.828	0.347

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